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FEASIBILITY TEST OF ELECTRODIALYTICALLY UPGRADED MSWI APC RESIDUE UTILIZATION IN MORTAR

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Abstract

Air pollution control (APC) residue from municipal solid waste incineration (MSWI) is classified as hazardous waste and disposed of, although it contains potential resources. Here, it was investigated whether MSWI APC residue may partly substitute cement in Portland cement mortar after upgrading by electrodialytic removal of heavy metals and salts. Electrodialytically upgraded MSWI APC residue was compared mainly to coal APC residue, which is used in Portland cement based materials already. Raw MSWI APC residue was also tested. The three APC residues were used in tests that included heavy metal (As, Ba, Cd, Cu, Cr, Pb and Zn) leaching and selected tests of Portland cement mortars with and without APC residue. The mortars were mixed with a w/b-ratio of 0.5, and 15 volume percent replacement of cement by APC residue and tested for heat development, compressive strength and possible cracks. The upgraded MSWI APC residue showed lower heavy metal leaching than the coal APC residue, except for Pb and Zn. The initial setting time was delayed about 10 hours in the mortar with upgraded MSWI APC residue compared to the reference mortar (no APC residue) and the coal APC residue mortar. The compressive strength of the mortars was tested after 7, 32 and 56 days and was in the order: reference >> upgraded MSWI APC residue > coal APC residue > raw MSWI APC residue. The results of this study indicated potential for using electrodialytically upgraded MSWI APC residue in Portland cement based materials.

1- INTRODUCTION

Air pollution control (APC) residue from municipal solid waste incineration (MSWI) is classified as hazardous waste due to its high content of salts and mobile heavy metals. Currently, APC residue from Danish MSWI is stabilized and disposed of. Contrarily, APC residue from coal combustion in Denmark is considered a valuable resource, which is reused in production of cement and as supplementary cementitious material in mortar and concrete. Similar to coal APC residue, the MSWI APC residue contains potential valuable recourses. And MSWI APC residue may be useful in the construction industry as secondary raw material, if the heavy metal toxicity is reduced. This may be done by removing the mobile fraction of the heavy metals from the MSWI APC residue by electrodialysis at the MSWI APC residue's alkaline pH.

The principle of electrodialysis (ED) is illustrated in figure 1. It is widely used e.g. for desalination of solutions in industrial scale. MSWI APC residues were subjected to ED in up to 10% solids containing suspensions, and the technique showed promising results for reduction of heavy metal mobility and removal of salts in laboratory scale [1]. The ED method was further developed to treat MSWI APC residue suspensions in both bench [2] and pilot scale [3] with promising results for removing both heavy metals and salts. The ED system consists of an ED stack with multiple concentrate compartments (concentrate) and compartments containing the APC residue suspension (diluate) in which both the concentrate and the diluate are pumped through the ED stack. The compartments are separated by ion exchange membranes and the setup is designed to remove anions and cations from the diluate to the concentrate by the applied current. There is a concentrate compartment next to both electrode compartments to limit the occurrence of other ions in the electrode compartments so the only electrode reaction is hydrolysis [3].

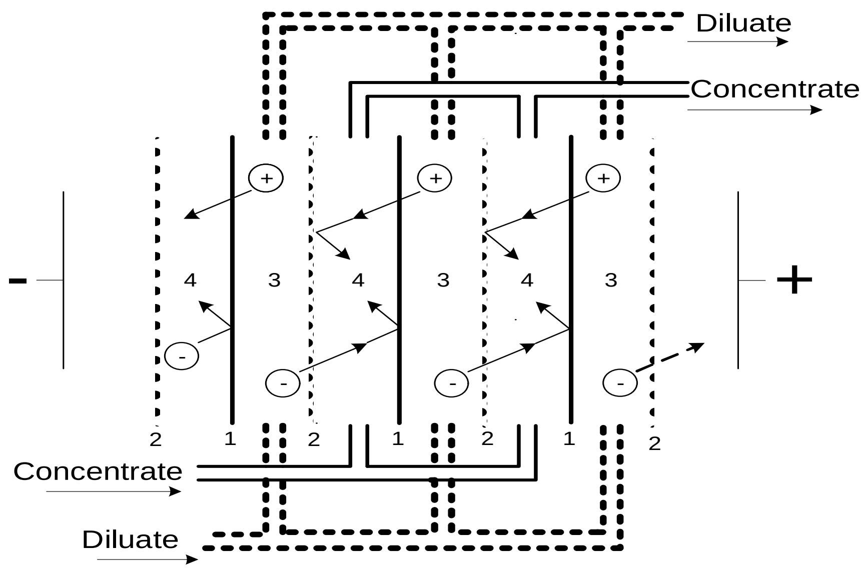

Figure 1: Principle of electrodialysis of APC residue.
1/2- ion exchange membranes, 3- diluate compartments, 4- APC residue suspension (diluate) compartments

Dewatering is required prior to reuse of the upgraded APC residue as a constituent in construction materials or for geotechnical purposes, while extracted salts and heavy metals may be reused and replace mining.

The aims of this study were 1) to compare the electrodialytically upgraded MSWI APC residue to coal APC residue and raw MSWI APC residue with regard to heavy metal leaching and 2) to investigate whether electrodialytically upgraded MSWI APC residue could replace some of the Portland cement in cement based materials and be used equally as coal APC residue.

2- MATERIALS AND METHODS

Materials

Raw MSWI APC residue after a dry flue gas cleaning process from a Danish waste incineration plant, REFA I/S, was used in this study. The raw MSWI APC residue was upgraded in an electrodialytic pilot scale experiment where up to 19 % of the total metal content was removed [3]. The coal APC residue was from Dong Energy A/S, Avedøreværket blok 1 and meets the requirements in EN 450 for reuse in concrete. Densities of the APC residues were determined by pycnometer according to DS/EN 196-6. For preparation of mortar low alkali sulfate resistant cement (CEM I 42.5 N) from Aalborg Portland and 0/4 mm quartz sand of class E from RN Sten & Grus, Hvidovre, Denmark were used.

Analytical

The APC residues were characterized for heavy metal content, leachable heavy metals and chloride content. Heavy metal concentrations (As, Ba, Cd, Cr, Cu, Pb, Zn) in the APC residue were measured by ICP-OES after pretreatment according to Danish Standard DS259. The units used in this paper are mg/kg for concentrations in dry matter. Chloride was measured by IC after extraction of 5 g APC residue with 25 mL distilled water overnight.

Leaching tests on the different APC residues were made according to CEN prEN 12457-3 part 1, with slight modifications. The liquid to solid (L/S) ratio was L/S 2, mixing 40 g APC residue and 80 ml distilled water. The suspension was shaken for 6 hours on an end-over shaker. pH was measured in the suspension before vacuum filtration through a 45 µm nucleo filter and the filtrate was acidified by addition of concentrated HNO_3 before measurement of heavy metals on ICP-OES.

Preparation of mortar

The mortar was mixed according to EN 196-1 at a water/binder-ratio of 0.5 and a sand/cement ratio of 3 (reference mix). The cement was substituted 15 % by volume with APC residue. Four experimental mortars were made: reference (no substitution), coal (substitution with coal combustion APC residue), raw MSWI (substitution with untreated MSWI APC residue), upgraded MSWI (substitution with electrodialytically upgraded MSWI APC residue). Compressive strength was tested after 7, 32 and 56 days on mortars prepared and cured according to DS/EN 12390-3 in triplicate for each mortar. Adiabatic heat development was measured after DS 423- 37 with slight modifications on one sample for each mortar continuously during 7 days after mixing. Possible cracks were investigated on fluorescence impregnated plane sections prepared from mortar bars cured for 28 days by Pelcon A/S, Denmark according to DS 423-39.

3- RESULTS AND DISCUSSION

Table 1 shows characteristics of the experimental APC residues. The heavy metal content was, as expected, significantly higher in the MSWI APC residues than in the coal APC residue. Even though a significant removal of heavy metals occurred during the electrodialytic treatment, the heavy metal content in the upgraded MSWI APC residue was higher than in the raw MSWI APC residue due to dissolution of APC residue during electrodialytic treatment [3]. The chloride content was significantly reduced during electrodialytic upgrading, but still higher than in the coal APC residue.

Parameter	Raw MSWI	Upgraded MSWI	Coal
As (mg/kg)	127	192	24
Ba (mg/kg)	370	448	1,100
Cd (mg/kg)	170	245	1.5
Cr (mg/kg)	93	150	46
Cu (mg/kg)	575	806	32
Pb (mg/kg)	2,220	2,150	19
Zn (mg/kg)	14,640	21,600	88
Cl (%)	16	0.5	0.02
Density (kg/m^3)	2860	1910	2160

Table 1: Characteristics of the experimental APC residues

The results of the leaching tests are found in table 2. They showed significantly reduced leaching of heavy metals from the upgraded MSWI, despite the higher total heavy metal content. The leached Cr concentration was however substantially higher in the coal APC residue, which may be used for construction materials without pretreatment. Leaching of Ba, Cd and Cr was also higher in the coal APC residue than the upgraded APC residue. An increased leaching of Cr in the upgraded MSWI APC residue compared to the raw APC residue should be investigated further, since Cr is difficult and important to stabilize in mortars [4]. Soluble Cr (VI) in cement can give eczema and EU has set a limit for cement to contain less than 2 mg/kg soluble Cr (VI) [5]. In this study, it was not investigated if Cr was present as Cr (VI) due to the difficulties of determination; however this should be included in future studies.

APC residue	pH	As	Ba	Cd	Cr	Cu	Pb	Zn
Raw MSWI	11.8	198	33,911	14	136	3291	484,450	60,459
Upgraded MSWI	12.3	<25	449	0.1	242	15	3,780	1,664
Coal	12.9	<25	669	79	1029	9	31	77

Table 2: Leached concentrations (µg/l) of heavy metals from the different APC residues.

The heat development of the mortars is illustrated in figure 2. The initial hydration was delayed in the mortars containing APC residue compared to the reference, but the highest heat acceleration period for all mortars occurred was within one day. The cumulative heat development measured within the first seven days was higher for all the mortars containing APC, which indicates that the APC either acted pozzolanic or induced a possible filler effect, see e.g. [6]. The initial setting times were found from the heat development curves, as the time where the extension of the linear part of the curve in acceleration period crosses the abscissa [7], and found to be: reference (3 h); coal (3-4 h); raw MSWI (9 h) and upgraded MSWI (13 h). The initial setting times were delayed when adding APC residue to the mortar, however to a lesser extent than a similar study with MSWI APC residue from a wet process [8]. This indicates a dependence on the composition of the APC residue. Taylor [9] claims that salts of Pb and Zn as well as phosphates can cause hydration retardation. Soluble Zn can be a hydration retarder because it can form amorphous layers on cement grains [10]. Pb has been observed to coat cement grains and precipitate on silicate surfaces [10]. It was seen that the observed setting times increased with increasing total Zn and Pb content. However, for the

MSWI APC residues, the raw APC residue with the highest leachable amount of Zn showed the shortest delay of the two, which suggests that the formation of $Zn(OH)_2$ is not the controlling mechanism. Addition of Cl to mortars can act as an accelerator or retarder depending on the concentration for the initial hydration and according to [11] as an accelerator at concentrations below 4 %. Here, the contrary occurred; adding the upgraded MSWI APC residue with a low Cl content (0.5 %) retarded hydration compared to the raw MSWI APC residue with high Cl content (16 %). At present the combined effect of soluble components on the setting time is not well described.

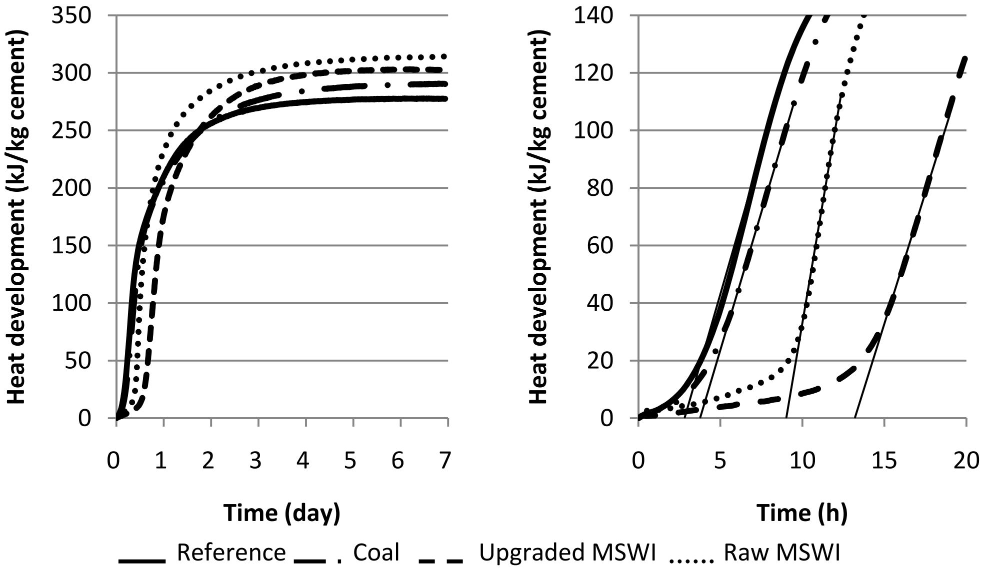

Figure 2: Accumulated heat of hydration in mortars containing APC residue

Compressive strength for the mortars are shown in figure 3 and the compressive strength generally increased with longer curing time.

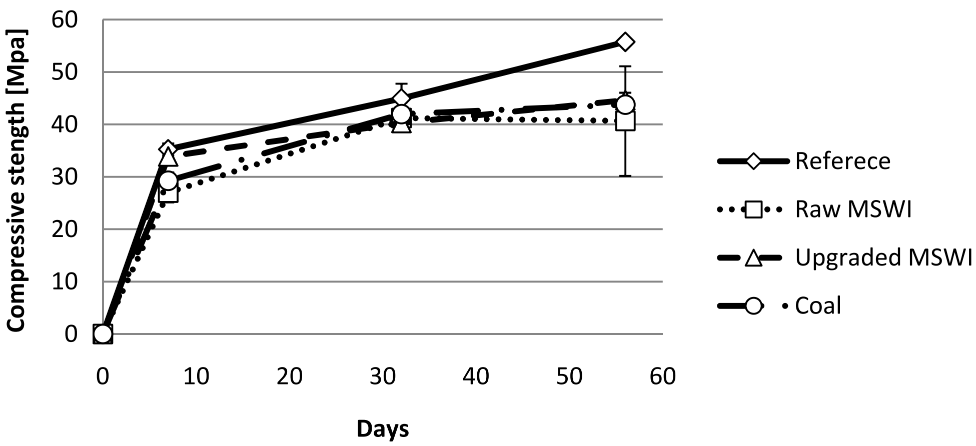

Figure 3: Compressive strength of the different mortars as a function of time with standard deviation bars

All the mortars containing APC residue exhibited lower compressive strength than the reference mortar, only the mortar containing upgraded APC residue showed similar strength properties after 7 days of curing. The results showed a larger strength increase from 32 to 56 days for the reference mortar, but limited strength change for the mortars containing APC residue. The compressive strength for the mortars with upgraded and coal APC residue were similar and only slightly higher than the compressive strength of the mortar with raw APC residue. Metallic Al and sulfate in MSWI APC residue is regarded as important factors to lower compressive strengths when added in mortars due to crack formation [4]. Contrary to [8] no visible crack formation was observed in the mortars in the study. During the mortar mixing, it was observed that the workability decreased when MSWI APC was added. The epoxy impregnated plane sections showed that the mortars with MSWI APC were slightly more heterogeneous than the reference and coal APC mortar. The calculated air content based on densities in the different mortar samples was between 10-12%, with the highest air content in the reference mortars. The visual air bubbles in the reference and coal APC mortars were more evenly distributed than in the MSWI APC mortars but similar in size and amount in all the mortars.

4- CONCLUSIONS

It was tested if electrodialytically upgraded MSWI APC residue could be used in Portland cement based materials. The heavy metal leaching was significantly reduced in the upgraded MSWI APC residue and for the Ba, Cd and Cr lower than from coal APC residue, which is already used as supplementary cementitious material in mortar and concrete. The upgraded MSWI APC residue was comparable to coal APC residue, and showed potential for reuse in Portland cement mortar. The compression strength of mortar containing upgraded MSWI APC residue was similar to mortar containing coal APC residue. However, the initial setting time was delayed in the mortar with upgraded MSWI APC residue compared to a reference mortar and coal APC residue.

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